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14. ABSTRACT The fracture and fatigue behavior of advanced refractory metals and silicide composites were investigated. Materials obtained from GE, Cabot, and WPAFB have been tested under both monotonic and cyclic conditions to measure the toughness and fatigue crack growth behavior. Comparison to some other refractory based systems is included.					
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**MICROSTRUCTURAL EFFECTS ON FRACTURE  
AND FATIGUE OF ADVANCED REFRACTORY  
METALS AND COMPOSITES**

**AFOSR F49620-00-1-0067**

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**FINAL TECHNICAL REPORT  
COVERING THE PERIOD 11/15/99 – 11/14/00**

**JUNE 2001**

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## Summary

While many advanced intermetallic systems possess desirable properties such as high temperature strength and stiffness, these systems typically do not exhibit adequate ductility and toughness at low temperatures. In addition, there is a need for the development of advanced processing techniques in order to provide materials in adequate quantities for subsequent use. The successful utilization of these desirable material properties also requires the development of techniques to impart higher toughness without sacrificing strength.

A variety of toughening mechanisms may be utilized to impart toughness in such systems. In brittle matrix systems, toughening may be achieved via the introduction of particles which are either brittle or ductile. In the former case, toughening may occur via crack bowing and/or deflection. The introduction of ductile particles may induce ductile phase toughening. The introduction of the ductile phases imparts some damage tolerance, the details of which have been modeled and experimentally investigated in our previous AFOSR grants. Much less work has focused on the behavior of such materials under cyclic loading conditions.

The one year AFOSR Program at Case Western Reserve University, AFOSR F49620-00-1-0067 focused on some key issues in the fatigue and fracture behavior of advanced intermetallic composite systems. Complementary work on the high rate fracture behavior and dynamic fracture toughness of Nb was also conducted. Close interaction and supply of materials from GE CRD, CSM, WPAFB, and Cabot, facilitated the work conducted.

The personnel supported over the period 11/15/99 – 11/14/00 are summarized below:

### Faculty:

Professor John J. Lewandowski

### Graduate Assistants:

Mr. Sergey Solv'yev	- M.S. Candidate	-	Partial Support
Mr. Deneesh Padhi	- M.S. Candidate	-	Full Support

### Undergraduate Students:

Mr. Jeff Showiak  
Mr. Robert Boyer  
Ms. Alwah Awadallah

The papers published under AFOSR support for the period 11/15/99 – 11/14/00 are summarized below:

1. Lewandowski, J.J. (1999). "Effects of the Addition of Toughening Ligaments on the Fatigue of Composites", In Fatigue '99, (X.R. Wu and Z.G. Zhang, eds.), EMAS 1998, Vol. III, pp. 1471-1477.
2. Zinsser, W.A., Solv'yev, S., and Lewandowski, J.J. (1999). "Fracture and Fatigue of Refractory Metal Intermetallic Composites", in Intermetallics VIII, Vol. 552, (E. George, M. Yamaguchi, and M. Mills, eds.), MRS, Pittsburgh, PA, pp. KK6.10.1-6.10.6.
3. Lowhaphandu, P., Montgomery, S.L., and Lewandowski, J.J. (1999). "Effects of Superposed Hydrostatic Pressure on Flow and Fracture of a Bulk Amorphous Alloy", *Scripta Metall. et Materialia*, **41(1)**, pp. 19-24.
4. Lowhaphandu, P., Ludrosky, L.A., Montgomery, S.L., and Lewandowski, J.J. (2000). "Deformation and Fracture Toughness of a Bulk Zr-Ti-Ni-Cu-Be Amorphous Alloy", *Intermetallics*, **8**, pp. 487-492.

## **Microstructural Effects on Fracture and Fatigue of Advanced Refractory Metals and Composites**

F49620-00-1-0067

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### **Program Research Objectives**

Our focus in this one year grant has been to determine the fundamental mechanisms controlling the fracture and fatigue behavior of very high temperature refractory metals (e.g. Nb, Mo) and intermetallic (e.g. Nb<sub>5</sub>Si<sub>3</sub>) materials toughened with ductile/tough phases. This research is utilizing materials processed at WPAFB from the binary Nb-Si system and alloy variants, as well as recently developed directionally solidified (DS) Nb-Ti-Hf-Si-Al-Cr (i.e. at GE CRD). Polycrystalline Mo processed at CSM has also been obtained. Relatively little is known regarding the monotonic fracture and cyclic fatigue behavior of the materials obtained from the binary Nb-Si systems as well as those obtained via alloying of both monolithic Nb, Mo, and composite Nb alloys. Recent work has begun to address these issues (1-25).

### **Research Approach**

1. Determine the microstructural factors controlling the fracture and fatigue behavior of the individual constituent phases that comprise toughened refractory metal intermetallic composite materials. The features (i.e. microstructure, stress state) controlling the impact and dynamic fracture toughness (i.e. K<sub>ID</sub>) of pure Nb and solid solution strengthened Nb-Zr and Nb-Si alloys were determined over a range of test temperatures. The desire was to determine whether cleavage fracture in such materials tested at very high strain rates obeys a critical tensile stress criterion. The level of dynamic fracture toughness possible when failure occurs via cleavage fracture was also found for polycrystalline Nb.
2. Continue to investigate the fatigue behavior of Nb toughened silicides over a range of test temperatures, R-Ratios, and alloy chemistries. Despite the great benefits afforded via the incorporation of ductile/tough Nb phases into brittle materials such as Nb<sub>5</sub>Si<sub>3</sub>, the behavior of such systems under cyclic conditions has only recently begun to be explored and very limited data exists. Fatigue tests have been conducted and all relevant portions of the fatigue crack growth curve have been developed including the threshold regime, the Paris Law regime, and the region of overload failure. Quantitative fractography of fractured specimens was utilized to rationalize the crack growth behavior.

Close contact has been maintained with researchers at WPAFB, UES, Inc., and GE CRD, where complementary studies are being conducted on the creep and oxidation resistance of materials processed via vacuum arc-casting/extrusion, vapor deposition, and directional solidification.

## Research Findings

### I. Dynamic Fracture Toughness ( $K_{ID}$ ) and Impact Toughness of Nb and Nb-Zr Monolithic Materials

#### a) Dynamic Fracture Toughness ( $K_{ID}$ ) of Monolithic Nb Materials

The present work has demonstrated that the dynamic fracture toughness of pure Nb (and Nb-1wt% Zr) is approximately  $40 \text{ MPa}\cdot\text{m}^{1/2}$  and relatively independent of test temperature over the range  $-196^\circ\text{C}$  to  $25^\circ\text{C}$ , as shown for pure Nb in Figure 1. Charpy impact specimens were fatigue precracked at  $-125^\circ\text{C}$  (in order to minimize the plastic zone size ahead of the fatigue precrack) prior to testing in an instrumented impact testing machine where the load-time trace during impact testing could be recorded. Changes in grain size from  $40 \mu\text{m}$  –  $165 \mu\text{m}$  did not significantly change the magnitude of the dynamic fracture toughness, as shown in Figure 1.

Such values are clearly important to determine since the fracture toughness of any composite that utilizes Nb as the toughening phase will predominantly rely on the toughness of that phase. It is clear that such materials can exhibit significant toughness despite the appearance of 100% cleavage.

Extensive fracture surface analyses were conducted in order to document the fracture mode and determine the location of possible fracture nucleation sites in the fatigue precracked impact toughness specimens. Matching surface fractography revealed the fracture mode to be transgranular cleavage. Furthermore, it was possible to locate the potential fracture nucleation sites by tracing the cleavage river lines back to suspected fracture origins as shown in Figure 2 for a fatigue precracked specimen tested in impact at  $-196^\circ\text{C}$ . In all cases, fracture nucleation occurred at distances ahead of the fatigue precrack consistent with the location of the peak tensile stress, indicative of tensile stress controlled cleavage fracture.

#### b) Notched Impact Fracture Behavior of Monolithic Nb Materials

The effects of changing grain size and test temperature on the Charpy impact energy were determined. As expected, increasing the grain size increased the ductile-to-brittle transition (DBTT) of notched polycrystalline Nb. However, little effect of grain size on the lower shelf impact toughness was obtained.

Extensive fracture surface analyses were conducted in order to document the fracture mode and determine the location of possible fracture nucleation sites in the notched impact toughness specimens. Matching surface fractography revealed the fracture mode to be transgranular cleavage for specimens tested below the DBTT. Furthermore, it was possible to locate the potential fracture nucleation sites by tracing the cleavage river lines back to suspected fracture origins. In all cases, fracture nucleation occurred at distances ahead of the notch consistent with the location of the peak tensile stress, indicative of tensile stress controlled cleavage fracture. The cleavage fracture stress calculated from the impact studies was consistent with our previous work under static conditions(16,17).

### **c) Fracture Behavior of Monolithic Mo Materials**

Initial materials have been obtained from CSM Industries where the processing conditions (i.e. rolling temperature, reduction ratio) of polycrystalline Mo have been changed to produce differences in grain size. Very little information exists on the fracture toughness of polycrystalline Mo in either unalloyed or alloyed conditions. Initial tests have been conducted on polycrystalline Mo that has been rolled at either 1450C or 1200C in order to vary the grain size and amount of retained work.

## **II. Fatigue Behavior of Nb-Si In-situ Composite Materials**

### **a) Fatigue Behavior of Nb-Si Composite Materials**

The cast/extruded composites exhibit a somewhat reduced threshold for fatigue in comparison to that of the monolithic alloys, although the values (e.g.  $\Delta K_{th} = 10 \text{ MPa-m}^{1/2}$ ) are still in the range of those exhibited by metallic materials (21). Furthermore, the values for the Paris law exponents,  $m$ , are also in the range of that exhibited by metallic materials (e.g. 3-5). The fatigue performance of the present materials are much better than that obtained on other intermetallics toughened with Nb/Nb alloys, where the fatigue performance of the composite was often not better than that of the semi-brittle intermetallic. Possible reasons for such differences are discussed elsewhere (19,21). Increasing the R-Ratio and changing the test temperature were shown to increase the rate of fatigue crack growth and lower the fatigue threshold, as shown in Figure 3.

Extensive quantitative fractographic analyses have revealed that for a given R ratio, increasing the level of  $\Delta K$  (and  $K_{max}$ ) produced a transition in fracture mode to increasing amounts of transgranular cleavagelike fracture of the primary Nb (19,20). Increasing the R-ratio increased the fatigue crack growth rate and this coincided with an increase in the amount of cleavage of the primary Nb. Tests conducted at -125C and 225C similarly showed an increase in the fatigue crack growth rate in comparison to that observed at room temperature. Quantitative fractography again revealed an increase in the amount of cleaved Nb in the tests conducted at -125C and 225C, consistent with the measured increase in crack growth rate. Higher temperature tests are planned.

In addition to the binary Nb-10Si composites, a number of multi component DS composites prepared by GE CRD have been tested (18,22). The fatigue performance of the DS composites is somewhat better (i.e. higher fatigue threshold, lower Paris law slope) than those of the cast/extruded materials over a range of R-ratios. Preliminary fractographic analyses reveal an absence of cleavage of the Nb present in the DS composites. Work is continuing in order to determine the mechanisms controlling fatigue in the DS materials because of the significantly different microstructures present in such materials.

### **Acknowledgment/Disclaimer**

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## References and Papers on Nb, Nb Silicide Acknowledging AFOSR Support

1. Lewandowski, J.J., Dimiduk, D., and Mendiratta, M. (1988). Proc. MRS Symp.-High Temperature Composites, (S. Fishman, et al eds.), Pergamon, Vol. 20, pp. 103-109.
2. Rigney, J., and Lewandowski, J. (1990). in Proc. 2<sup>nd</sup> Int'l. Ceramic Sci.Tech. Cong.-Adv.Composite Matls, Westerville, OH., (M.Sacks, et. al. eds.), pp. 519-525.
3. Mendiratta, M., Lewandowski, J.J., and Dimiduk, D. (1991). Metallurgical Transactions A, 22A, pp. 1573-84.
4. Rigney, J.D., Singh, P.M., and Lewandowski, J.J. (1992), JOM, Aug. 1992, pp. 36-41.
5. Kajuch, J., Rigney, J., and Lewandowski, J. (1992). Matls Sci. Eng., A155, pp. 59-65.
6. Kajuch, J., Rigney, J.D., and Lewandowski, J.J. (1993). , in Proc. MRS Symp. (J.D.Whittenberger, et al eds.), MRS, Pittsburgh, PA, Vol. 288, pp. 853-860.
7. Zhang, J., and Lewandowski, J.J. (1994). Jnl of Materials Science, 29, pp. 4022-4026.
8. Lewandowski, J.J. (1994). in In-situ Composites: Science and Technology, (M. Singh and D.Lewis, eds.), TMS, Warrendale, PA, pp. 159-167.
9. Short, J.W., Kajuch, J., and Lewandowski, J.J. (1994), in Proc MRS Symp-Intermet. Matrix Composites III, (J.A. Graves, et al eds.), MRS, Pittsburgh, PA, pp. 285-293.
10. Rigney, J.D., Singh, P.M., and Lewandowski, J.J. (1994), in MRS Proceedings-Vol 332, MRS, Pittsburgh, PA, pp. 503-509.
11. Kajuch, J., Short, J.W., and Lewandowski, J.J. (1995). Acta Met., 43, pp. 1955-1967.
12. Mendiratta, M.G., Goetz, R., Dimiduk, D.M., and Lewandowski, J.J. (1995). , Metall. Trans. A, 26A, pp. 1767-1777.
13. Rigney, J.D., and Lewandowski, J.J. (1995), in Fatigue and Fracture of Ordered Intermetallic Matls II, (T. Srivatsan, et al eds.), TMS, Warrendale, PA, pp. 339-359.
14. Lewandowski, J.J. and Rigney, J.D. (1996), in Proc. NATO ASI-Mech. Behavior Matl's at High Temp., (C. Moura Branco eds.), Kluwer Dordrecht, pp. 535-545.
15. Rigney, J.D., and Lewandowski, J.J. (1996). , Metall. Trans. A., 27A, pp. 3292-3306.
16. Samant, A., and Lewandowski, J.J. (1997). Metall. Trans. A., 28A, pp. 389-399.
17. Samant, A., and Lewandowski, J.J. (1997). Metall. Trans. A, 28A, pp. 2297-2307.
18. Bewlay, B.P., Lewandowski, J.J., and Jackson, M.R. (1997), JOM, August, pp. 44-47.
19. Lewandowski, J.J. and Zinsser, W., (1998). Metall. Trans. A., 29A, pp. 1749-1757.
20. Zinsser, W. and Lewandowski, J.J. (1998). Scripta Metall. 28, 12, pp. 1775-1780.
21. Lewandowski, J.J. (1998). in Fatigue '99, (X.R. Wu, ed.) EMAS, 1999, pp. 1471- 1477.
22. Zinsser, W.A., Solov'yev, S. and Lewandowski, J.J. (1999) in Intermetallics VIII, Vol. 552, (E. George, et al eds.), MRS, Pittsburgh, PA, 1999, pp. KK6.10.1 – KK6.10.6.
23. Lowhaphandu, P. and Lewandowski, J.J. (1999), Metall. Trans. A., 30A, pp. 325-334.
24. Lowhaphandu, P. and Lewandowski, J.J. (1998), "Fracture Toughness and Notch Toughness of Bulk Metallic Glass", Scripta Metall, 38, 12, pp. 1811-1817.
25. Zwonitzer, S.A., Rozak, G. and Lewandowski, J.J. (1998), in Mo and Mo Alloys, (A. Crowson, et al eds.) TMS-AIME, Warrendale, PA, pp. 111-125.
26. Lowhaphandu, P., Montgomery, S.L., and Lewandowski, J.J. (1999). "Effects of Superposed Hydrostatic Pressure on Flow and Fracture of a Bulk Amorphous Alloy", Scripta Metall. et Materialia, 41(1), pp. 19-24.
27. Lowhaphandu, P., Ludrosky, L.A., Montgomery, S.L., and Lewandowski, J.J. (2000). "Deformation and Fracture Toughness of a Bulk Zr-Ti-Ni-Cu-Be Amorphous Alloy", Intermetallics, 8, pp. 487-492.

## Awards Received:

Leonard Case Professorship in Engineering – Effective July, 2000.  
 CTSC Technical Educator Award – Cleveland Technical Societies, awarded May 2000.  
 Nominee – Carl F. Wittke Award for Undergraduate Teaching, May 2000.  
 Institute of Metals (UK) Charles Hatchett Award for work on Nb, Awarded 1999/2000.  
 MRS Undergraduate Student Research Award – Robert Boyer (2000).

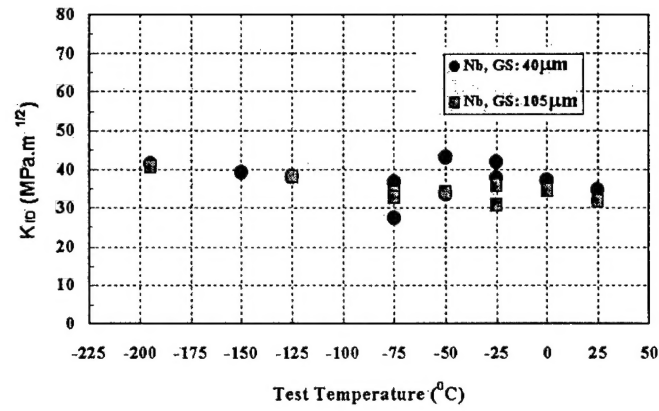
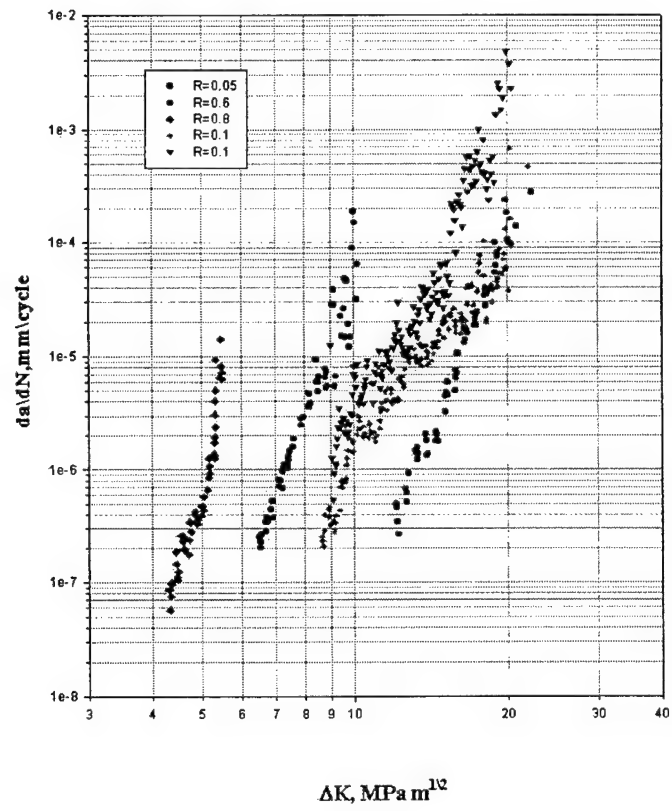


Figure 1. Dynamic Fracture Toughness as a Function of Test Temperature



Figure 2. Fracture Surface Montage of an Nb<sub>Cp</sub> 40 μm Fatigue-Precracked Charpy Specimen Tested at -196°C. Apparent Cleavage Fracture Nucleation Sites are Shown Inside the Boxes



**Figure 3.** Fatigue crack growth rate of Nb-10 Si Composites as a Function of  $\Delta K$  for different R-ratios.

**REVIEW PAPER SUBMITTED TO ISSI-III**

# FLOW, FRACTURE, AND FATIGUE OF Nb AND Nb SILICIDE INTERMETALLIC COMPOSITES

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## Abstract

The flow, fracture, and fatigue behavior of refractory metal toughened intermetallic composites is significantly affected by the mechanical behavior of the refractory metal phase. This paper reviews some of the balance of properties obtained in composites based on the Nb-Si system. Since some of the composite properties are dominated by the behavior of the refractory metal phase, the paper begins with a review of data on monolithic Nb and its alloys. This is followed by presentation of results obtained on Nb-Nb silicide composites and a comparison to the behavior of some other high temperature systems.

## Introduction

The potential use of alloys and intermetallic composites based on refractory metals continues to receive attention for some of the highest temperature structural applications (1,2). While the oxidation resistance of monolithic refractory metals are generally poor, the intermetallic compounds of refractory metals with silicon (e.g. Nb, Mo) are of interest because of their high melting points, their increased oxidation resistance, and their relatively low densities (1). In particular, alloys/composites in the systems Mo-Si-B-X (X = W, V, Cr, Nb, Al, Ge, Re) and Nb-Ti-Hf-Cr-Al-Si-Ge combine many of the attractive features of refractory metals and intermetallic compounds. Despite the potential of silicides for use as high temperature materials such as turbine airfoils or some hypersonic applications for service temperatures ranging from 1000-1400°C, relatively little is known about their structure and properties (in monolithic or composite form) at ambient or elevated temperatures. Some of the property requirements for the hottest parts of aircraft engines are provided elsewhere (2).

Predominant issues in the mechanical behavior of these advanced materials are their ambient and high temperature properties, including the strength, ductility, toughness, creep, and fatigue performance, while the oxidation resistance at intermediate and high temperatures is also under investigation. Recent works have successfully demonstrated the potential of improving the toughness of niobium silicide systems via compositing with ductile Nb phases utilizing ingot casting and extrusion, directional solidification, as well as lamination via PVD or powder metallurgy techniques (1-4). Parallel efforts are being developed to improve the oxidation and creep resistance of Nb via alloying without compromising the toughness. Much less is known about the effects of such alloying on creep, and in particular, the fatigue behavior of the niobium silicide based system. Features controlling the fracture and fatigue performance of the Mo-Si-B-X systems have been studied far less than that of the Nb based systems.

The flow, fracture, and fatigue performance of the toughened Nb silicide based composites are significantly affected by the flow, fracture, and fatigue performance of the toughening phase. In

most of the composite systems studied to date, the Nb toughening phase contains additional alloying elements, either due to contamination (e.g. O, C, etc.) or due to alloying (e.g. Si, Hf, Ti, Cr, Ge, etc.) to improve the oxidation resistance and/or other properties. In order to be able to document the effects of toughening phase(s) on the composite behavior, it is necessary to understand the effects of microstructural changes on the flow, fracture, and fatigue performance of the toughening phase itself. As such, this paper will first review the available literature on the factors which control the flow, fracture, and fatigue behavior of the monolithic toughening phases based on Nb. The effects of microstructural changes on the behavior of polycrystalline Nb will be covered first, followed by recent fracture and fatigue studies over temperatures ranging from 77K - 773K on Nb toughened Nb silicide composites prepared either by arc casting/extrusion or directional solidification (DS). The presence of the brittle silicide intermetallic in the composite may produce constrained flow and elevated tensile stresses in the Nb phase(s), thereby changing the fracture behavior in the Nb toughening phase. As such, the behavior of unconstrained and constrained Nb will be presented for a range of temperatures prior to the discussion of the composite behavior. Test temperatures as low as 77K will be covered as this should illustrate the effects of changes in stress state and flow behavior on the flow/fracture of the toughening phase itself.

## Polycrystalline Nb Flow and Fracture Behavior

The flow stress of polycrystalline Nb and its alloys are relatively insensitive to changes in grain size over the range 5  $\mu$ m to roughly 500  $\mu$ m. Figure 1 presents a compilation (5) of 0.2% offset yield strength data over a range of temperatures and grain sizes taken from the literature as well as from work conducted at CWRU. It is clear that relatively minor increases in strength result from significant changes in the grain size. The Hall-Petch slope obtained for changes in grain size ranging from 20  $\mu$ m to roughly 165  $\mu$ m was  $8.7 \times 10^4 \text{ N/m}^{-3/2}$  (5), consistent with previous work on polycrystalline Nb (6-8) shown in Figure 1.

The Ultimate Tensile Stress (UTS) is also relatively insensitive to changes in grain size as shown in Figure 2 (5).

Figures 3 and 4 show the effects of changes in test temperature on the yield stress and UTS, respectively (5). The strength of commercial purity Nb at 298K is 150-300 MPa and at 77K can exceed 1 GPa, while still exhibiting significant ductility and ductile fracture (5). Nb with solid solution additions of Si is significantly strengthened, with strengths at 298K exceeding 350 MPa while still possessing some ductility (8-10). Reductions in test temperature to 77K further increase the yield strength to near 1 GPa, although the fracture at 77K occurs via cleavage with low ductility (8-10). The strength of commercial purity Nb is quite low at high temperatures, while Figure 5 shows the increase in strength for the Nb-Ti-Cr-Al system over a range of temperatures (11-14). Some of the Nb silicide based composites are also shown in Figure 5 and illustrate that

strengths approaching 1 GPa at 298K are possible, with high temperature strengths well in excess of that of the monolithic toughening phase (11-14).

The Ductile to Brittle Transition Temperature (DBTT) of polycrystalline Nb is dependent on processing condition and chemistry as well as grain size. Literature data summarized in Figure 6 (5) reveals an effect of processing condition as well as grain size on both the DBTT as well as the notched impact energy. Decreases in grain size reduce the DBTT and increase the impact energy, while high purity Nb prepared via electron beam melting (EBM) exhibited the lowest DBTT and highest notched impact energy for the conditions reported in Figure 6 (5). Work conducted at CWRU on commercial purity Nb heat treated to different grain sizes similarly reveals an effect of grain size on the DBTT, Figure 7 (5), which shows the amount of brittle (i.e. cleavage) vs ductile (i.e. non-cleavage) fracture present on fractured impact specimens tested at different temperatures. For example, at 248K, coarse grained Nb exhibited 75% cleavage fracture while the fine grained Nb exhibited 0% cleavage.

The competition between flow and cleavage fracture in polycrystalline Nb and Nb alloys (including Si-solid solution strengthened Nb) was investigated (6,8,9) where the cleavage fracture stress was measured via notched specimens. The results of those works (6,8,9) revealed that cleavage fracture in polycrystalline Nb and Nb alloys occurs by reaching a temperature independent critical cleavage fracture stress. The magnitude of the cleavage fracture stress was shown to depend solely on the grain size in the materials studied, with decreases in grain size producing increases to the cleavage fracture stress (6). The temperature independent values for cleavage fracture stress varied from 1000–1500 MPa depending on the grain size (6,8,9). Extensive fractography of the notched bend specimens revealed that cleavage fracture initiated ahead of the notch in the region of peak tensile stress (6,8,9), entirely consistent with classic theories of tensile stress controlled cleavage fracture. The high constraint and stress levels present in the composites shown in Figure 5 indicate that cleavage of the refractory phase may occur in such systems.

The static fracture toughness of polycrystalline Nb has been determined on fatigue precracked bend specimens tested over temperatures ranging from 77K to 298K (15). In addition, dynamic fracture toughness tests,  $K_{ID}$ , have been determined on fatigue precracked Charpy impact specimens tested in an instrumented impact machine (5). The latter are shown in Figure 8 and reveal that the fracture toughness obtained under dynamic loading conditions is in the range 30-40 MPa-m<sup>1/2</sup> from 77K to above 298K, while fracture surface examination revealed the fracture to consist of 100% cleavage fracture (5). The values obtained for the static fracture toughness over a more limited temperature range were in close agreement with the data obtained under impact conditions (6), suggesting that the minimum fracture toughness of the polycrystalline Nb samples is quite large (i.e. 30-40 MPa-m<sup>1/2</sup>), despite the appearance of 100% cleavage fracture. The fracture toughness of the Nb-Si solid solution alloy was approximately 30 MPa-m<sup>1/2</sup> (8,9) at 298K.

The fatigue crack growth behavior of polycrystalline Nb has not been investigated to any great extent. Recent work (16-18) has revealed thresholds for fatigue in the range of 10-12 MPa-m<sup>1/2</sup> at R=0.1 at 298K. Paris Law slopes were in the range 2-4, consistent with most metallic materials. Increases in stress ratio decreased the fatigue threshold somewhat without significantly

changing the Paris Law exponent, consistent with most metallic materials. This is shown in Figure 9 and includes data for some composites to be discussed below.

### Fracture and Fatigue Behavior of Nb-Nb Silicide Composites

Composites based on the binary Nb-10 a/oSi system shown in Figure 10 have been extensively investigated. The majority of the work (1-4, 8-10, 16-18) on this system has been conducted on arc cast/extruded material, subsequently heat treated 1500C/100 hrs in order to equilibrate the structure to contain Nb<sub>5</sub>Si<sub>3</sub> and Nb(ss-Si). The structure shown in Figure 10 exhibits elongated primary Nb(ss-Si) present at about 50 volume %. The primary Nb has a grain size of roughly 25-50 μm, with the size of the primary Nb roughly 50-100 μm. Secondary Nb is present at about 25 volume % with a size of about 1 μm. The remaining 25% of the structure is the brittle Nb<sub>5</sub>Si<sub>3</sub>.

Fracture toughness tests conducted on the monolithic Nb silicide revealed that it possessed a fracture toughness of only 1-2 MPa-m<sup>1/2</sup> (10). Fracture toughness tests on the Nb-Si composite shown in Figure 10 were conducted so that crack growth occurred perpendicular to the extrusion direction. R-curve behavior and significant increases in toughness were exhibited by the Nb-Si composite due to the toughening provided by the Nb(ss-Si). Figure 11 presents a sequence of photographs taken at increasingly higher load (i.e. A-B-C-D) in a test conducted at 298K (8). Cracks which have impinged on the Nb are blunted and the Nb deforms significantly before fracture. Tests over a wider range of loading rates are summarized in Figure 12 (8). The toughness, roughly 24 MPa-m<sup>1/2</sup> is relatively insensitive to large changes in loading rate and test temperature. The very high levels of toughness exhibited at 77K at the fastest loading rates occurred with a preponderance of cleavage of the primary Nb in the composite (8). Despite the presence of cleavage fracture of the primary Nb, the fracture toughness far exceeded that of the Nb silicide. This appears to be consistent with the high toughness of the polycrystalline Nb presented earlier (cf Fig. 8) despite the appearance of 100% cleavage. Testing at 773K does not significantly change the fracture toughness of the Nb-10 a/o Si composite or that of a variety of Nb-Ti-Si directionally solidified (DS) composites (19) despite a change in fracture mode of the toughening phase over that range of temperatures.

Exposure of the Nb-10 a/oSi system to oxygen at high temperature produced significant hardening and embrittlement of the Nb(ss-Si), thereby reducing the fracture toughness of the composite (20). Recent alloying studies have produced significant increases to the oxidation resistance without significantly degrading the fracture and fatigue performance (2), as will be discussed below.

Few studies of the fatigue crack growth behavior of toughened Nb-Si composites have been conducted. Early work (21-23) on Nb toughened intermetallics suggested that the fatigue performance of such toughened systems might be as poor as that of the monolithic brittle matrix, with Paris Law slopes approaching 100 in some cases. In those cases, the toughening constituent failed prematurely via fatigue (21-23). In order to address the relevance of these issues for the Nb-Si system, extensive testing over a range of stress ratios and test temperatures has been conducted (16-18, 24), followed by quantitative fractography in order to document the type of fracture mechanism(s) operating in the different fatigue regimes

for the Nb-10 a/o Si composite shown in Figure 10.

Recent fatigue crack growth experiments at 298K on the Nb-10 a/o Si system (16-18, 24) are summarized in Figure 13. Fatigue crack growth experiments were conducted on bend bar specimens tested at stress ratios ranging from 0.05 – 0.8 on a closed loop MTS servo-hydraulic testing machine. Crack growth was monitored via the use of foil resistance gages (i.e. KRAK-gages) bonded to the outer surface of the specimens. Multiple tests were conducted for each stress ratio and representative results are shown in Figures 9 and 13. The fatigue threshold and Paris Law slope at  $R=0.1$  is similar to that of metallic materials as shown in Table 1 and Figure 9. Figures 9, 13, and Table 1 also reveal that increasing the stress ratio decreases the fatigue threshold and increases the Paris Law slope. While the former observation (i.e. decreased fatigue threshold) is consistent with the behavior of metals (25), the increase in Paris Law slope with increasing stress ratio is typically not observed in monolithic metals. Initial work (16,17) revealed that cleavage fracture of the primary Nb occurred in the Paris Law regime of the Nb-10 a/o Si composites. Recent tests conducted over a wider range of stress ratios combined with quantitative fractography have determined the amount of cleaved primary Nb at different  $\Delta K$  in Figure 13, as discussed below (24).

Figure 14 illustrates the effects of changes in  $\Delta K$  on the amount of cleaved primary Nb (16-18, 24). It is clear that increases in  $\Delta K$  increase the amount of cleaved primary Nb. However, as shown earlier, cleavage of Nb is typically considered a static fracture mode controlled by reaching a critical value of the cleavage fracture stress (6). If this is the case, cleavage during fatigue crack growth should be controlled by  $K_{max}$  in the fatigue loading cycle. The data in Figure 14 are replotted in Figure 15 using  $K_{max}$  instead of  $\Delta K$  (16,17,24). The use of  $K_{max}$  appears to normalize all of the data obtained at different stress ratios at 298K and suggests strongly that the increase in the Paris Law slope exhibited in Figure 13 and Table 1 with increasing stress ratio is due to the intervention of static modes of fracture (i.e. cleavage) in the primary Nb. This further suggests that microstructure manipulation and/or testing at different temperatures could affect the tendency for cleavage of the primary Nb, and hence significantly change the fatigue crack growth characteristics. Preliminary work of this nature has produced fatigue crack growth characteristics and fractographic observations which are consistent with these predictions (24).

Higher alloy variants of the Nb-10 a/o Si system have been prepared in order to improve the oxidation resistance (2). Both arc-casting/extrusion as well as DS materials have been prepared. Table 1 also summarizes the fatigue threshold and Paris Law slope of the materials prepared and tested to date (18,24). It is again clear that these Nb alloy toughened composites can exhibit fatigue thresholds, Paris Law exponents, and values for fracture toughness which are metallic-like in character. In order to demonstrate this, Figures 9 and 16 were prepared from the data obtained presently as well as that taken from the literature (25, 26). Included in Figures 9 and 16 are data for ceramics, toughened ceramics, ceramic composites, and preliminary work on Mo based intermetallic composites. The fatigue thresholds are as low as  $1.5 \text{ MPa}\cdot\text{m}^{1/2}$  at  $R=0.1$ , with Paris Law slopes for these other systems approaching 60 - 100 in some cases (25, 26), in contrast to the high threshold and low Paris Law slope (i.e.  $<10$ ) values generally exhibited by the Nb-Si system at  $R=0.1$ . Although the Nb toughened Nb Silicide composites exhibit an

increase in Paris Law slope with increased stress ratio, this is apparently due to the intervention of static modes of fracture during the fatigue test. Reducing the tendency for the Nb phase(s) to cleave via microstructure changes, or testing at higher temperatures, should further improve the Paris Law slope of the Nb-Si systems.

### Summary

The data obtained to date indicates that it is also important to investigate the behavior of the monolithic toughening phase(s) in order to understand their role in the toughening and fatigue behavior of the composites. Further work is clearly necessary in order to demonstrate that alloying additions are effective in improving the oxidation resistance without negatively impacting the properties. In addition, there is virtually no data on the high cycle fatigue and small crack growth behavior of such systems. Additional fatigue tests at different temperatures and stress ratios are also needed to determine the generality of the arguments presented here.

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### References

1. DM Dimiduk, MG Mendiratta, and PR Subramanian, in ISSI-I Structural Intermetallics, (R Darolia et al eds.), TMS, Warrendale, PA, (1993) 619-630.
2. BP Bewlay, JJ Lewandowski, and MR Jackson, JOM, August, (1997) 44-47.
3. MG Mendiratta, JJ Lewandowski, and DM Dimiduk, Metall. Trans. A, 22A (1990) 1573-1583.
4. MG Mendiratta and DM Dimiduk, Metall. Trans. A, 24A (1993) 501-504.
5. D Padhi, Ph.D. Thesis, Case Western Reserve University, Dept. Materials Science and Engineering, January 2001.
6. A Samant and JJ Lewandowski, Metall. Trans. A, 28A (1997) 389-399.
7. MA Adams, AC Roberts, and RE Smallman, Acta Metall., 8 (1960) 328-337.
8. JD Rigney and JJ Lewandowski, Metall. Trans. A, 27A (1996) 3292-3306.
9. MG Mendiratta, R Goetz, DM Dimiduk, and JJ Lewandowski, Metall. Trans. A, 26A (1995) 1767-1777.
10. J Kajuch, JW Short, and JJ Lewandowski, Acta Metall. et Materialia, 43 (1995) 1955-1967.
11. PR Subramanian, MG Mendiratta, DM Dimiduk, and MA Stucke, Mater. Sci. Eng., A239-240 (1997) 1.
12. BP Bewlay, MR Jackson, and HA Lipsitt, Metall. and Materials Trans. A, 27A (1996) 3801.
13. MR Jackson and KD Jones, in Refractory Metals: Extraction, Processing, and Applications, (KC Liddell, DR Sadoway, and RG Bautista, eds.), TMS, Warrendale, PA, (1990) 335.
14. MG Mendiratta, JJ Lewandowski, and DM Dimiduk, Metall. Trans. A, 22A (1991) 1573-1583.
15. A Samant and JJ Lewandowski, Metall. Trans. A, 28A (1997) 2297-2307.



16. WA Zinsser and JJ Lewandowski, Scripta Metall. et Materialia, 38(12) (1998) 1775-1780.
17. WA Zinsser and JJ Lewandowski, Metall. Trans. A, 29A (1998) 1749-1757.
18. JJ Lewandowski, in Fatigue '99, (XR Wu and ZG Zhang, eds.), Volume III, EMAS (1999) 1471-1477.
19. JD Rigney, BP Bewlay, DM Dimiduk, and JJ Lewandowski, Unpublished Research, (1998).
20. JD Rigney, PM Singh, and JJ Lewandowski, JOM, August (1992) 36-41.
21. KTV Rao and RO Ritchie, Mater. Sci. Eng. A153 (1992) 479-485.
22. KTV Rao, WO Soboyejo, and RO Ritchie, Metal. Trans. A, 23A (1992) 2249-2257.
23. WO Soboyejo and SML Sastry, Mater. Sci. Eng. A171 (1993) 95-104.
24. S Solv'yev and JJ Lewandowski, Unpublished Research, Case Western Reserve University, Cleveland, OH (2000).
25. RO Ritchie, Intl Journal Fracture, 100 (1999) 55-83.
26. H Choe, D Chen, JH Schneibel, and RO Ritchie, in Fatigue and Fracture of High Temp. Mat'l's, (PK Liaw, ed.), TMS Warrendale, PA (2000) 17-24.

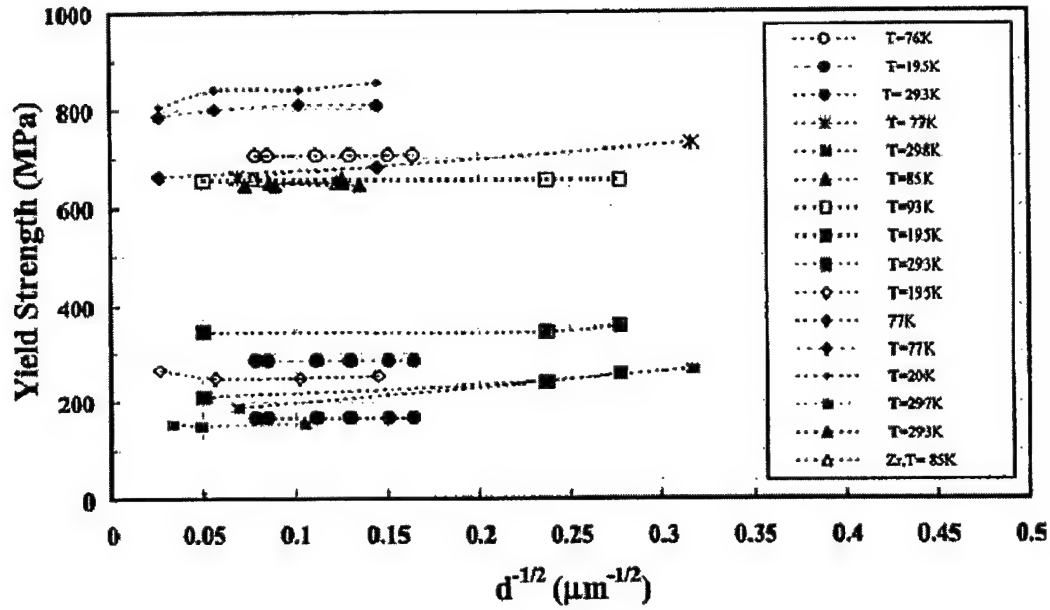


Figure 1. Effects of Changes in Grain Size on Yield Strength of Polycrystalline Nb and Nb alloys (5).

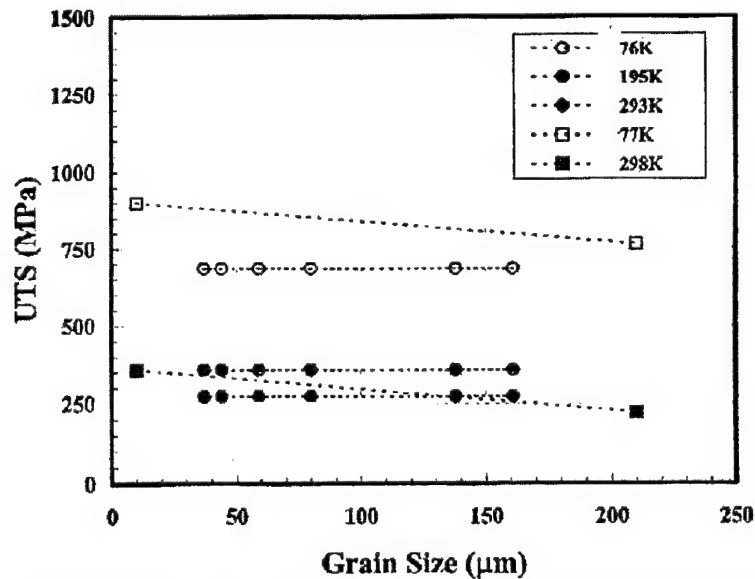


Figure 2. Effects of Changes in Grain Size on UTS of Polycrystalline Nb and Nb alloys (5).



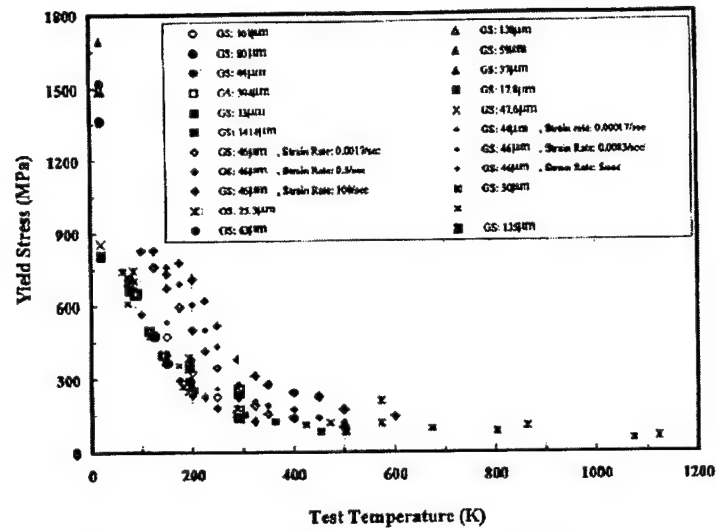


Figure 3. Effects of Changes in Test Temperature on Yield Stress of Nb and Nb alloys (5).

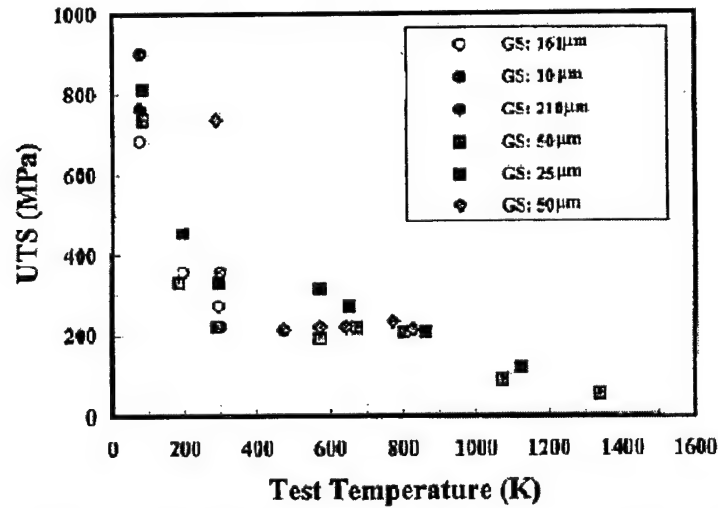


Figure 4. Effects of Changes in Test Temperature on UTS of Polycrystalline Nb and Nb alloys (5).

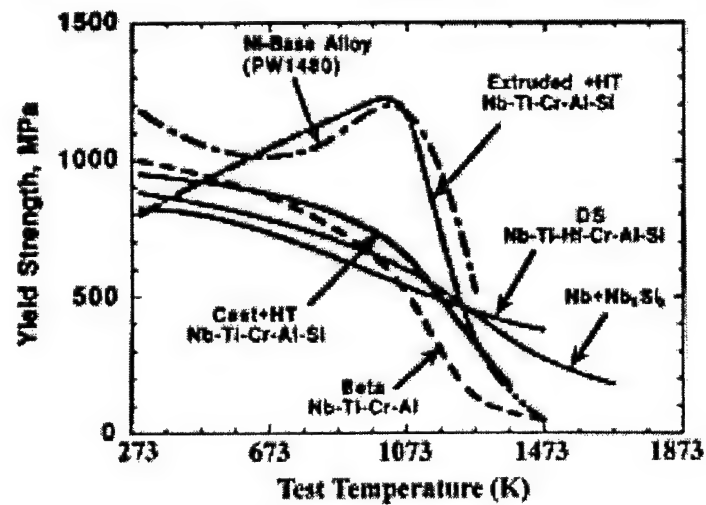


Figure 5. Effects of Test Temperature on Strength of Monolithic Nb Alloys and Nb Silicide Composites (11-14).

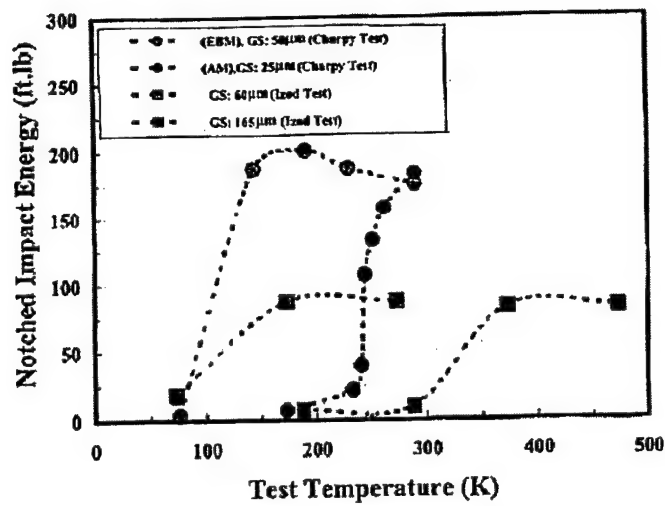


Figure 6. Effects of Changes in Grain Size and Processing Conditions on Charpy Impact Energy and DBTT of Nb (5).

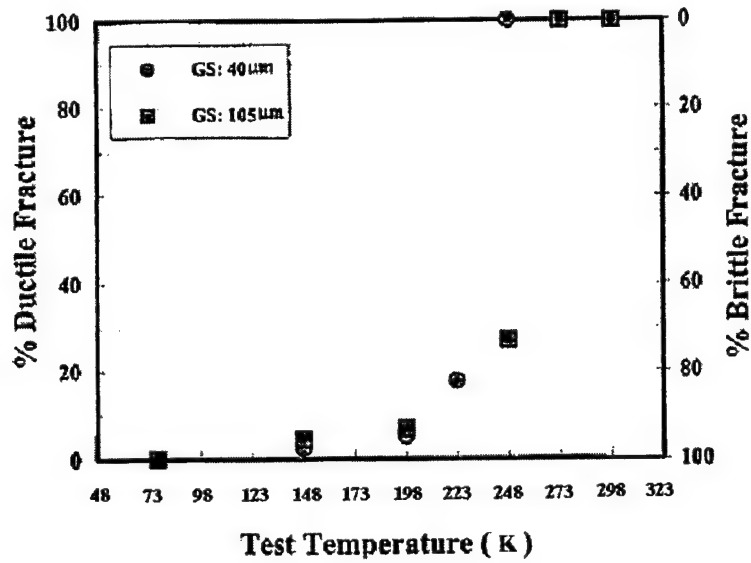


Figure 7. Effects of Changes in Grain Size on DBTT and Fracture Surface Appearance of Nb (5).

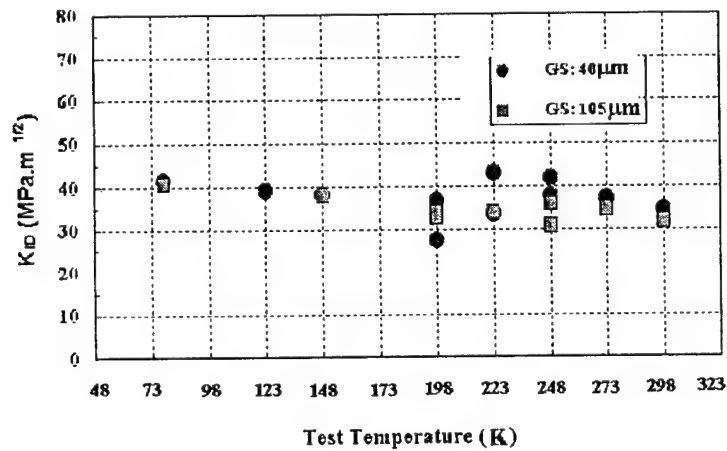


Figure 8. Effects of Changes in Grain Size and Test Temperature on Dynamic Fracture Toughness,  $K_{ID}$ , of Nb (5).

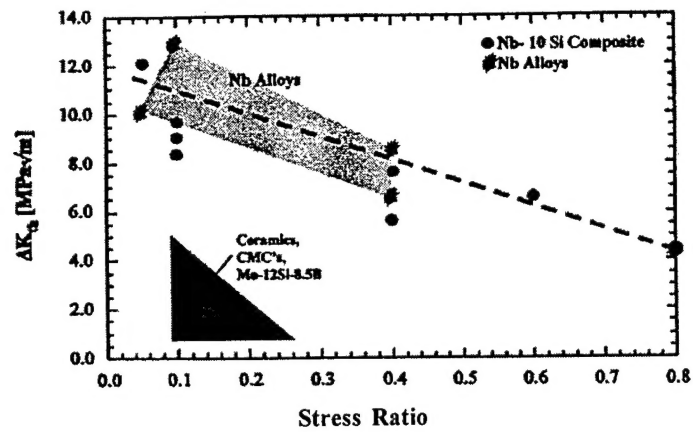


Figure 9. Summary of Effects of Changes in Stress Ratio on Fatigue Threshold of a Variety of High Temperature Materials Tested at 298 K (16-18, 24-26).

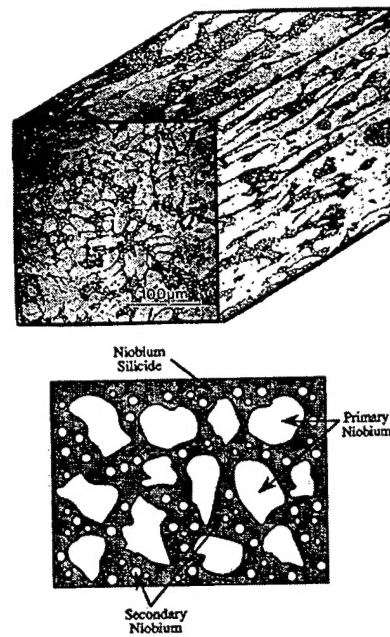


Figure 10. Typical Microstructure of Arc Cast/Extruded Nb-10 at.o Si Alloy (3,8).

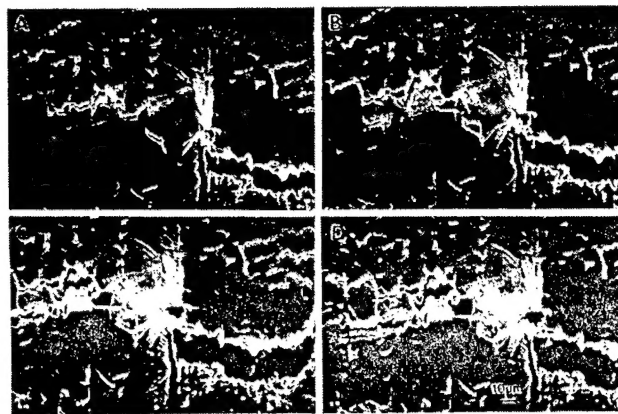


Figure 11. In-situ Fracture Experiment Showing Crack Impingement on Tough Nb in Nb-10 at.o Si Composite. Increasing load from A-D (8, 20).

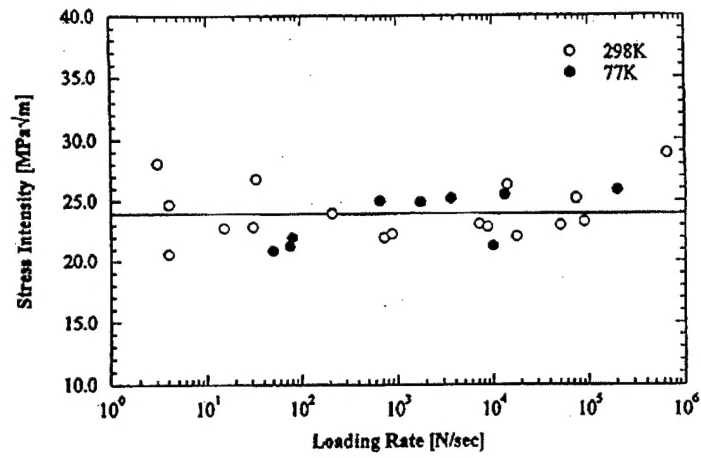


Figure 12. Effects of Changes in Loading Rate and Test Temperature on Toughness of Nb-10 a/o Si Composite (8).

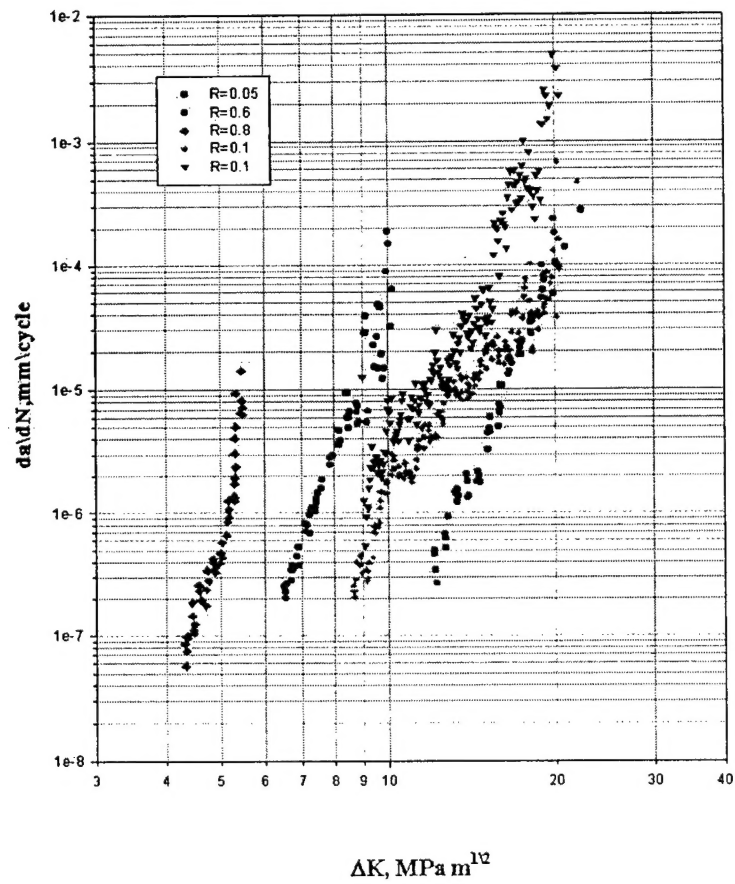


Figure 13. Effects of Changes in Stress Ratio on Fatigue of Nb-10 a/o Si Composites.

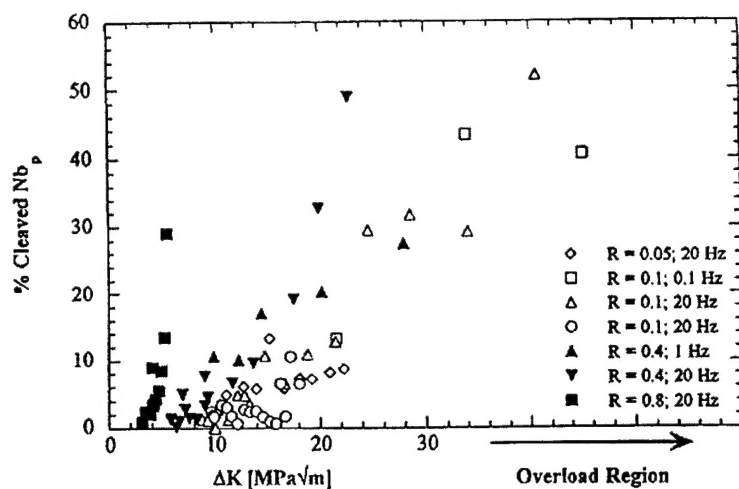


Figure 14. Quantitative Fractography Showing Increase in Cleaved Primary Nb with Increasing  $\Delta K$  in Nb-10 a/o Si Composites.

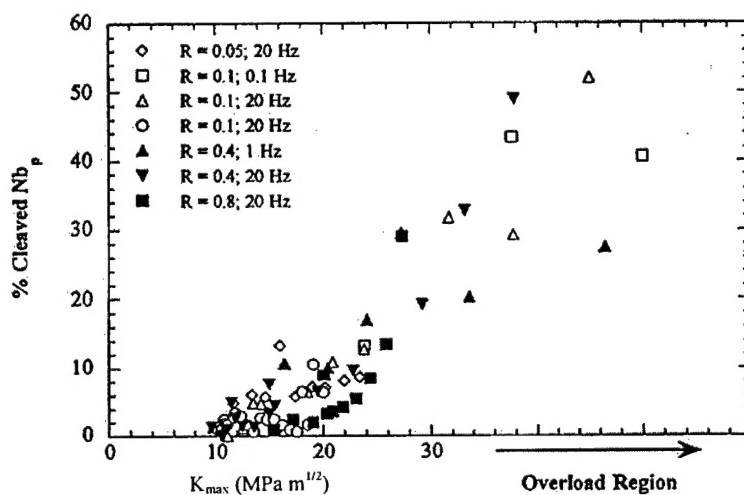


Figure 15. Quantitative Fractography Showing Increase in Cleaved Primary Nb with Increasing  $K_{max}$  in Nb-10 a/o Si Composites.

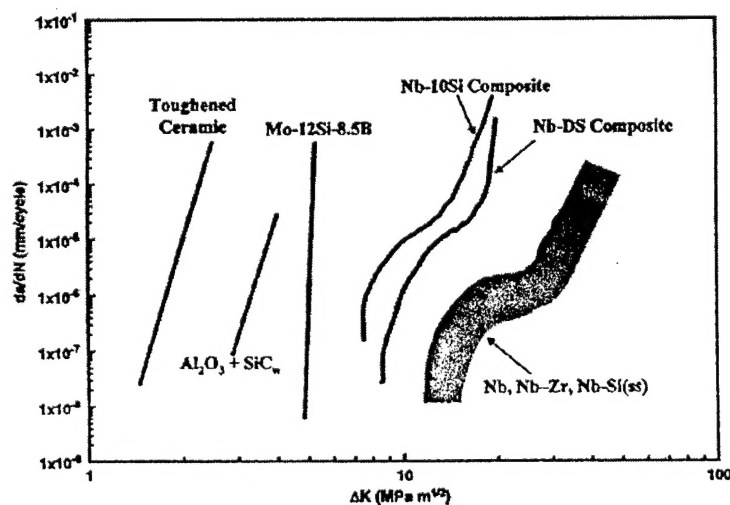


Figure 16. Comparison of Fatigue Behavior at 298 K (16-18, 24-26).

TABLE I-- FATIGUE CRACK GROWTH BEHAVIOR  
OF Nb-Nb SILICIDE COMPOSITES

Materials	Processing Condition	R-ratio	$\Delta K_{th}$ , MPa-m <sup>1/2</sup>	m-Paris Law Slope	$K_{IC}$ , MPa-m <sup>1/2</sup>
Nb-10Si	Extruded + HT	0.05	12.0	6.1	23.4*
	Extruded + HT	0.1	9.0	6.6	24.1*
	Extruded + HT	0.1	8.4	6.5	24.3*
	Extruded + HT	0.1	9.7	6.3	22.3*
	Extruded + HT	0.6	6.5	8.9	25.4*
	Extruded + HT	0.6	6.0	9.9	24.8*
	Extruded + HT	0.8	3.2	21.6	25.2*
	Extruded + HT	0.8	4.4	16.9	27.2*
Nb-15Si	Extruded +HT	0.1	4.4	6.4	20.6*
	Extruded +HT	0.4	4.1	-	-
	Extruded +HT	0.6	4.0	5.6	22.5*
	Extruded +HT	0.8	3.7	-	24.9*
Nb-12Si	DS	0.1	13.2	16.7	18.1
Nb-18.2Si	DS	0.1	2.5	-	3.3
Nb-30Ti-8Cr-10Al-14Si	DS	0.1	-	-	8.3**
Nb-42.5Ti-15Si	DS	0.1	5.5	9.7	10.2
Nb-Ti-Hf-Cr-Al-Si	DS	0.1	8.5	2.9	24.2
Nb-22Ti-3Hf-2Cr-2Al-17Si	Extruded +HT (1500°C/100hr)	0.1	7.1	4.8	24.4
		0.4	5.6	4.1	19.8
Nb-22Ti-3Hf-2Cr-2Al-17Si	Extruded +HT (1400°C/100hr)	0.1	7.2	4.8	17.8
		0.4	3.9	4.2	13.9

\* $K_{max}$  at fatigue overload    \*\*Without precracking